

# Lawrence Berkeley National Laboratory

## Recent Work

**Title**

Understanding Heterogeneous Chemical Processes Using X-ray Techniques.

**Permalink**

<https://escholarship.org/uc/item/2pf2t7mc>

**Journal**

Accounts of chemical research, 49(1)

**ISSN**

0001-4842

**Author**

Kevan, Steve

**Publication Date**

2016

**DOI**

10.1021/acs.accounts.5b00542

Peer reviewed

## Understanding Heterogeneous Chemical Processes Using X-ray Techniques

Stephen D. Kevan

Over the past few decades, advances in synchrotron radiation technology have provided increasingly incisive X-ray tools that are now essential to probe problems in nearly all areas of chemical research, ranging from aerosols to zeolites. X-ray scattering and diffraction, for example, are now routinely used to determine the structure of complex and imperfectly ordered supramolecular materials, biopolymer complexes, and nanoporous membranes. X-ray spectroscopy offers excellent chemical and orbital contrast to probe operating catalysts, batteries and fuel cells, diverse environmental and geological systems, and many more. The high flux and pulsed nature of synchrotron radiation is now regularly combined with X-ray scattering and spectroscopy to provide powerful probes of chemical kinetics and dynamics with atomic and molecular sensitivity. X-ray microscopies are being combined with this growing toolbox to probe chemical, electronic, and orbital morphologies with nanometer resolution.

The commissioning of much brighter free electron laser and diffraction-limited storage ring X-ray sources in the next few years will further embellish these tools, and in particular will revolutionize our ability to understand diverse heterogeneous systems. For example, we envisage the ability to measure spontaneous spatiotemporal chemical kinetics in nanoporous and nanostructured media with nanometer and nanosecond precision, supporting unique applications across a broad range of modern chemical research.

The present ACR Focus Issue provides examples of what is presently possible in chemical research using synchrotron radiation and glimpses of what will become possible in the near future. Readers are encouraged to think about how the results discussed in these accounts will be impacted by emerging tools. For example, the account by Pollock and deBeer describes how X-ray spectroscopy can be used to understand the electronic structure of transition metal centers. In the future it will be possible to do that with nanometer resolution in highly dispersed heterogeneous catalysts. Combining such spectroscopy measurements with simultaneous atomic structure determination will achieve an enduring goal: closing the structure-function loop in catalysis. Similar ideas apply to an environmental context in the account by Toner, et. al, which examines the chemistry that occurs on environmental nanoparticles, and to biochemical systems like Photosystem II, which is already under intense study using these techniques. Two of the accounts, by Stoerzinger, et. al., and by Knop-Gericke, et. al., discuss a modern variant of X-ray photoelectron spectroscopy that is performed at a pressure of a few mbar, called ambient pressure XPS. Applications have grown rapidly over the past decade; for example, it is now possible to study functioning electrochemical systems with XPS under a thin layer of water. The new sources will make it possible to image functioning

catalytic and electrocatalytic systems with composition and nanostructure designed to optimize activity and selectivity. The final account by Gessner and Guehr examines new techniques to measure the motion of charge in space and time, through molecules and junctions. Emerging X-ray capabilities at free electron laser sources will unravel the important role of nanometer-scale heterogeneity in these photochemical charge transfer processes.

It has been said (probably incorrectly) that 20<sup>th</sup> century technology was based on homogeneous systems, but that 21<sup>st</sup> century technology will focus on designing heterogeneous structures to accomplish chemical and physical processes that operate with high efficiency and selectivity. Much of that transition will be based on improved understanding of how interfaces and interphase regions operate in real devices. The tools to accomplish that will need to combine nanometer spatial resolution and high chemical contrast with enough signal to allow studies on relevant time scales. Clearly no single kind of tool will provide a complete solution, but ongoing improvements the brightness of X-ray sources is rapidly improving the ability of X-ray tools to probe functioning heterogeneous systems in new and important ways.